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INVESTIGATION OF A CESIUM IODIDE PHOTODISSOCIATION  
LASER AS A SOURCE AT 4555 NM(U) NAVAL OCEAN SYSTEMS  
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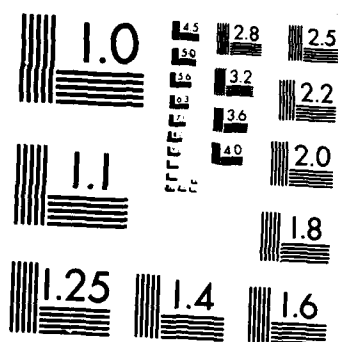
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**Technical Report 1136**  
September 1986

# Investigation of a Cesium Iodide Photodissociation Laser as a Source at 455.5 nm

Frank Hanson

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## **ADMINISTRATIVE INFORMATION**

This report describes experiments designed to generate a spectrally narrow laser source. The work was carried out by the Electro-Optics Devices Branch (Code 843).

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19 ABSTRACT (Continue on reverse if necessary and identify by block number) This investigation is concerned with both the feasibility of generating a spectrally narrow laser source at 455.5-nm by photodissociation of cesium iodide, and the possibility of amplifying this output in a parametric process using a suitable nonlinear material. Experiments using a 193-nm ArF laser as a photodissociation source are described. Only very weak output at 455.5-nm was observed, perhaps due to competition from other lines which depleted the 7P3/2 state and the eventual buildup of the ground 6S1/2 state. The lines at 917-nm were observed to be super-radiant at a pumping energy on the order of 1 mJ/cm <sup>2</sup> .			
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## OBJECTIVE

Determine the feasibility of generating a spectrally narrow laser source at 455.5 nm by photo dissociation of cesium iodide. Examine the possibility of amplifying this output in a parametric process using a suitable nonlinear material.

## RESULT

Generating a spectrally narrow and stable laser source by dissociating alkali halides is unsuitable for this application.

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## 1. INTRODUCTION

This project is concerned with the generation of a spectrally narrow laser source at 455.5-nm coincident with the atomic cesium  $7p_{3/2}-6s_{1/2}$  transition. Such a laser, together with an atomic cesium filter/receiver based on this transition (reference 1), would be useful in a laser communications system able to operate over long distances with a background of sunlight. It would be very attractive to have a laser based on the identical transition used in the detection process since the operating wavelengths would be inherently matched. This would avoid all problems associated with wavelength stability and drift that are a concern with alternative laser sources for this filter/receiver. Of course, this laser would not be tunable and, therefore, no means would exist to compensate for a Doppler wavelength shift between source and detector.

In the late 1970s, Ehrlich and Osgood (references 2,3) investigated a large class of alkali-halides that could be photodissociated using rare gas-halide excimer lasers resulting in excited neutral alkali atoms. They found many of the transitions in the excited alkali manifolds to be super-radiant. In particular, cesium iodide could be photodissociated with the 193-nm output of an ArF laser and would give super-radiant emission at several lines in the near infrared. There was some indication that the 455-nm line originating from the excited  $7p_{3/2}$  cesium state was also weakly super-radiant. Because this transition terminates on the ground  $6s_{1/2}$  level, any super-radiance (or laser action) would be very sensitive to any mechanism that would lead to a buildup of the ground state. An energy level diagram for the excited states of cesium is shown in figure 1.

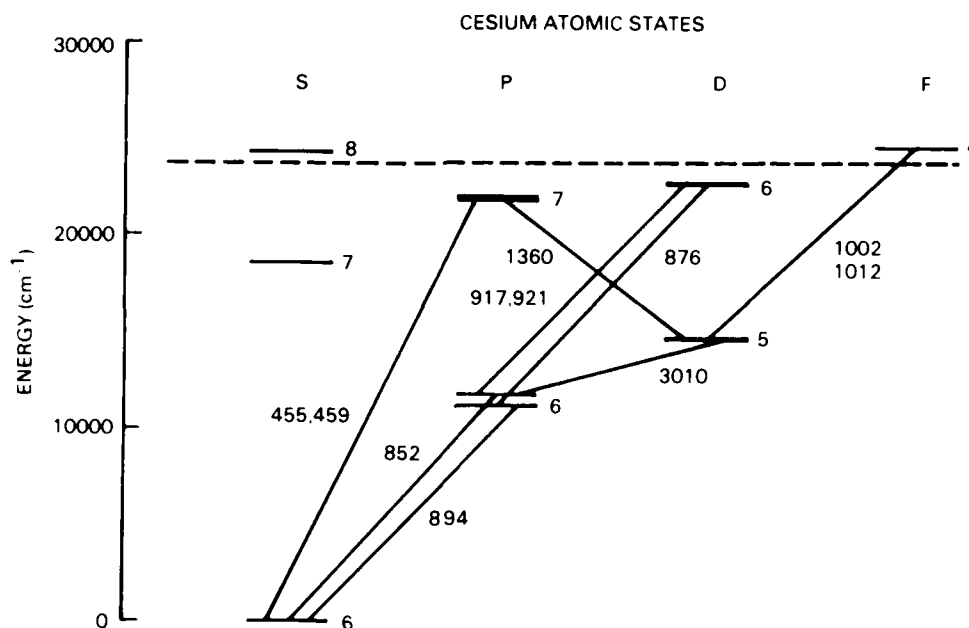


Figure 1. Energy levels for atomic cesium with transition wavelengths in nm. The energy reached by dissociating cesium iodide into ground state iodine  $2P_{3/2}$  with 193 nm light is shown (dashed line).

The absorption in the ultraviolet by the alkali halides shows distinct bands corresponding to unbound or very weakly bound excited molecular states which then generally dissociate into a ground state  $2P_{3/2}$  halogen atom and an excited alkali atom (reference 4). At 193-nm, the excess energy above the dissociation energy of CsI to ground state fragments is about  $24300\text{ cm}^{-1}$  (reference 5). This is nearly coincident with the  $8S_{1/2}$  Cs state at  $24317\text{ cm}^{-1}$  (reference 6). The energy defect to the  $7P_{3/2}$  state is over  $2000\text{ cm}^{-1}$ . Photodissociation to the ground state of cesium is very low following absorption at 193-nm because there is no nearby excited iodide state available to take up the excess energy. Ehrlich and Osgood (reference 3) estimate the relative dissociation yield for the  $7P_{3/2,1/2}$  states to be on the order of 15 percent (reference 2).

Competition from other lines originating from the 7p states, buildup of the ground state from both radiative and nonradiative quenching processes, and spectral linewidth are all concerns that would have to be investigated to determine the potential usefulness of this approach. Also, the possibility of using an electric discharge in place of the pump laser source must be considered. Direct electric discharge excitation is difficult from a practical standpoint due to high temperatures and corrosive environments. An inductively coupled microwave discharge might be feasible, however, electronic excitation, in general, would not selectively excite the higher energy states and, therefore, would not be expected to produce an inversion for transitions to the ground state. This approach cannot be completely discarded however. Neiger et al. (reference 7) have recently reported inversions of several resonance transitions in atomic thulium by electron collisions in a TmI3 discharge cell. They estimate a relatively weak electron impact cross section for dissociation into the thulium ground state to explain their results.

Although the overall efficiency for this type of laser operating at 455-nm would most likely be low even under optimistic assumptions, it would possibly still be useful if the output could be amplified to a resonable power level by an efficient parametric process using an efficient pump laser. The nonlinear parametric process would allow poor beam quality in the pump (within limits set by various phase matching restrictions) while maintaining the high spectral purity and beam quality of original signal.

The investigation consists of characterizing a small CsI photodissociation laser, evaluating the potential for operation at 455-nm and, finally, determining and evaluating possible materials for parametric amplification using either a 308-nm XeCl laser or the 355-nm third harmonic of a Nd:YAG laser as a pump source.

## 2. THE EXPERIMENT

A 12-inch tube furnace with an independently heated side chamber, capable of temperature in excess of 800 C, was constructed. An approximately 18-cm quartz cell was designed and fabricated with suprasil grade ultraviolet (UV) quartz windows fused on at Brewster's angle. A sidearm allowed evacuation and filling. The empty cell was initially pumped on while being flamed to red hot with a natural gas torch. A small quantity of cesium iodide (99.999 percent, Aldrich Company) was placed inside and the cell was again pumped on at 250 C for 2 hours after which the sidearm was fused closed.

The optical layout of the experiment is shown schematically in figure 2. A Tachisto 400X excimer laser operating with ArF was used to longitudinally pump the Csl cell. During experiments with a rear high reflector for 455-nm, a UV-grade prism was placed in the pump path between the cell and the mirror. With this arrangement the 455-nm mirror would not have to transmit 193-nm, which is a difficult requirement. Quartz discs (tilted slightly) were used to attenuate the pump for energy in/out measurements. A beam splitter placed immediately before the oven allowed the pump intensity to be monitored with a fast (1-2 ns) S-5 vacuum photodiode. The pump energy measurement was calibrated with a Gentec pyroelectric detector. Various filters were placed after the cell to eliminate the transmitted UV and allow attenuation of the signal. The signal, in a small solid angle along the pump axis, was analyzed using a 0.5 meter monochromator and EG&G PAR model 1452 silicon photodiode array (OMA) or RCA 8852 or 7102 photomultipliers and Tektronix 7912 digitizer with a fast 7A19 amplifier. Relative wavelength response of the detection system was made using a quartz halogen lamp operating at a known temperature. Absolute energy calibration of the Csl output was done in a very approximate manner by estimating the collected solid angle from the lamp and using a very large monochromator slit width.

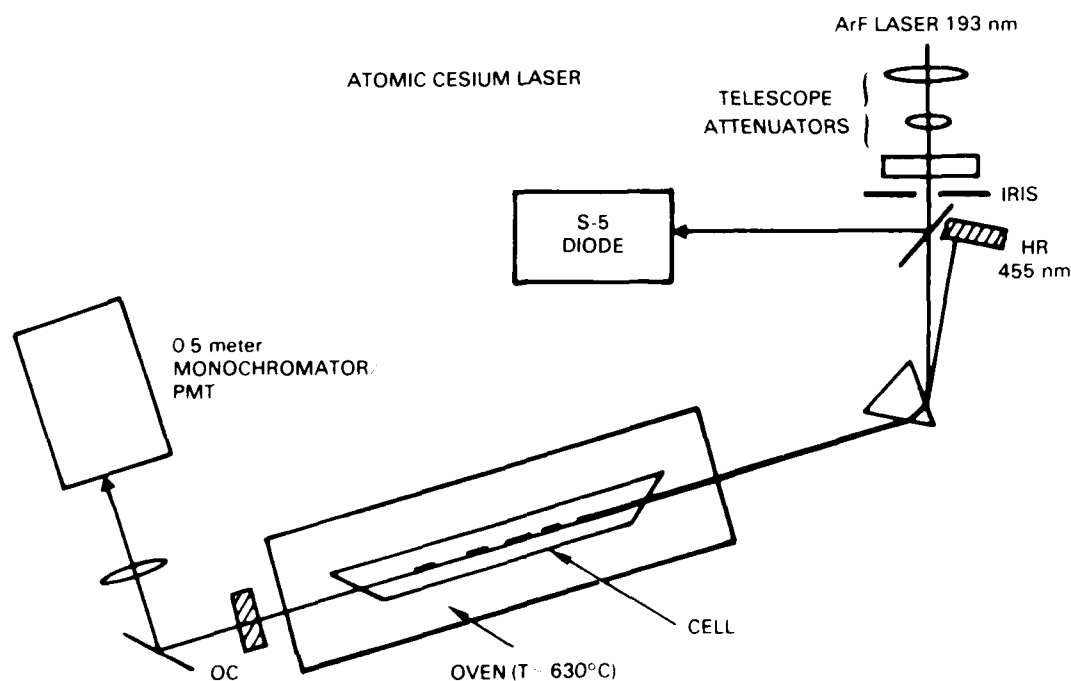


Figure 2. Experimental setup showing intracavity Brewster prism. The low temperature sidearm is not shown on the the cell.

### 3. RESULTS

The spectral output from the photodissociated CsI is shown in figure 3 using a coarse diffraction grating (500-nm blaze) and oma. Here the cell is at 705 C and the sidearm at 632 C. No correction for the wavelength response of the detection system was made. However a cesium discharge lamp spectrum is also shown for comparison in figure 4. The excited state distribution is not expected to be very similar to that found in the CsI cell, but some interesting observations can be made. First, the dominant features in the lamp spectrum are the two strong resonance line emissions at 852-nm and 894-nm. Secondly, the 917-nm, 876-nm, and 921-nm lines all originate from the 6d state and show roughly comparable intensities from the lamp. Out of the CsI cell the 917-nm line is 4 to 5 times stronger than the 876-nm line and the line at 921-nm is lost in the wing of the 917-nm line. The relative strength of the emission at 917-nm is clear evidence of super-radiance. Also, since the 852-nm and 894-nm lines are suppressed from the CsI cell, it seems likely the ground 6s state is significantly populated. There is, perhaps, an indication of the 455-nm line in second order in the wing of the intense 917-nm line. This line is clearly visible in the lamp spectrum.

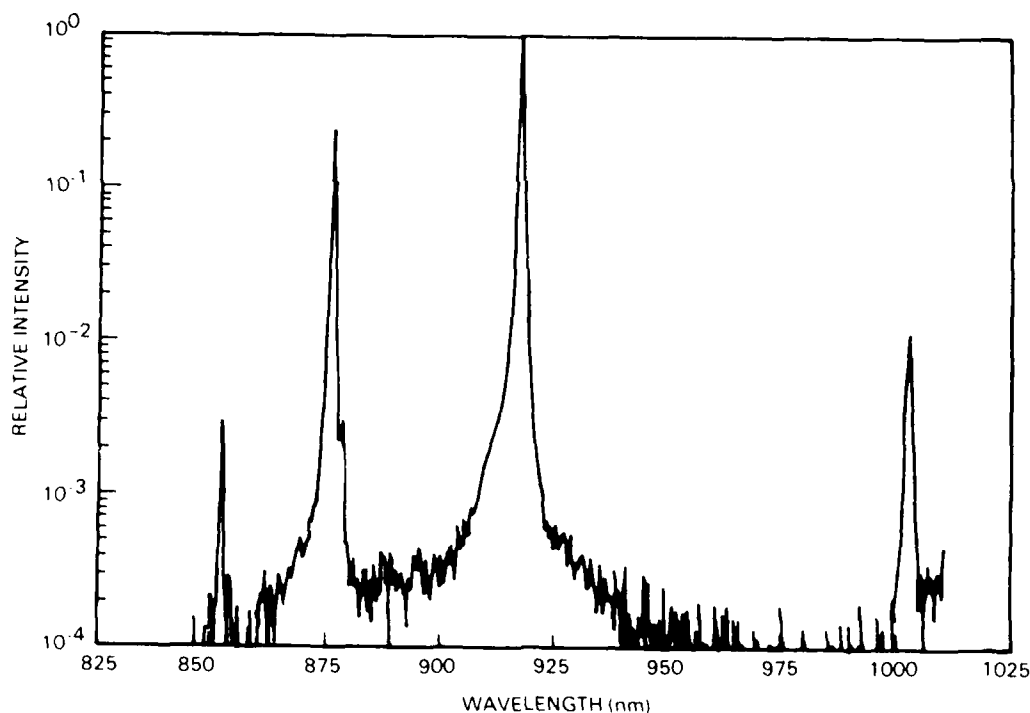


Figure 3. Relative spectral emission from cesium iodide cell uncorrected for detection response.

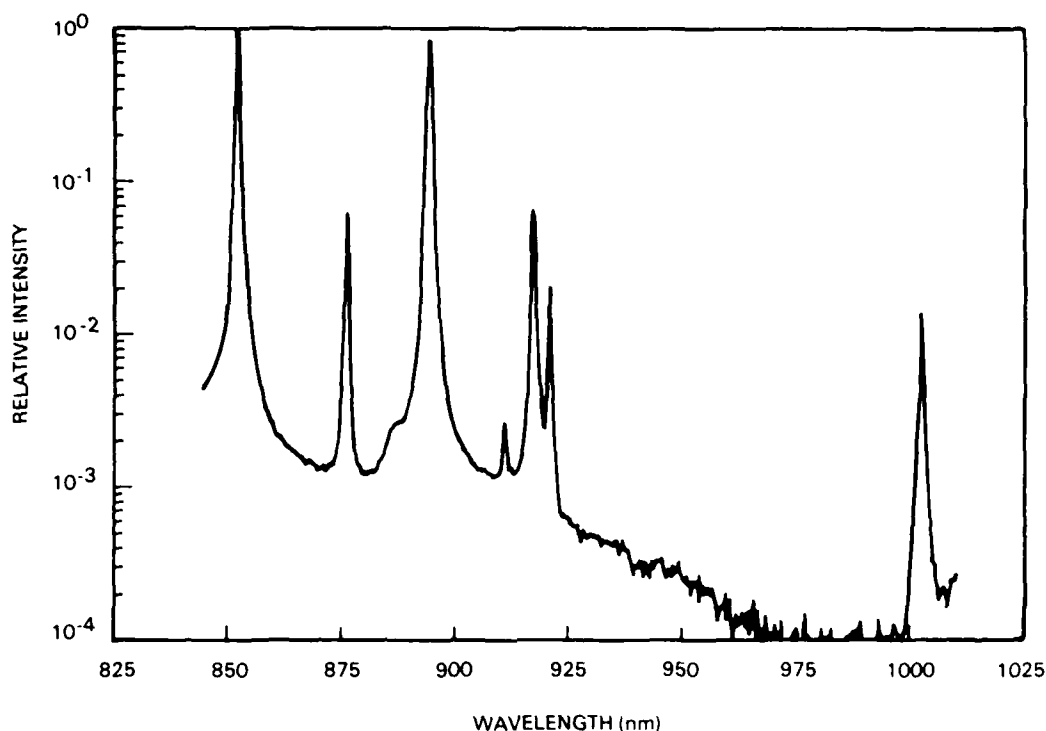


Figure 4. Relative spectral emission from a cesium lamp using the same instrumentation as in figure 3.

The output at a single wavelength was measured with the monochromator-photomultiplier combination. Figure 5 shows typical time traces of the 193-nm pump and the cell output at 455-nm. The optimum sidearm temperature for maximizing 455-nm output was found to be about 630 C (figure 6). Subsequent measurements were taken at this temperature with the cell somewhat hotter (approximately 700 C) to prevent condensation at the Brewster windows. Vapor pressure data for Csl is given in reference 8 and the calculated number density is shown in figure 7. The decrease in output at higher temperatures may be due to increased quenching from Csl or multistep ionization leading to quenching and ground state production. Formation of dimers increases at higher temperature but should still be small at these temperatures (reference 9). At 630 C the saturated vapor pressure of Csl is about 0.1 Torr (reference 8) which gives a density of about  $10^{15}/\text{cc}$  at 700 C. Using an absorption cross section of  $1.2 \times 10^{-16} \text{ cm}^2$  at 193-nm, estimated from measurements taken down to 200-nm (reference 4), results in an absorption path length of about 9 cm. The cell path length is twice this value. The pumping aperture was about  $0.2 \text{ cm}^2$  giving a volume of 3.6 cc containing  $3.6 \times 10^{15}$  Csl molecules. This would require 3.7 mJ of 193-nm energy absorbed for complete dissociation.

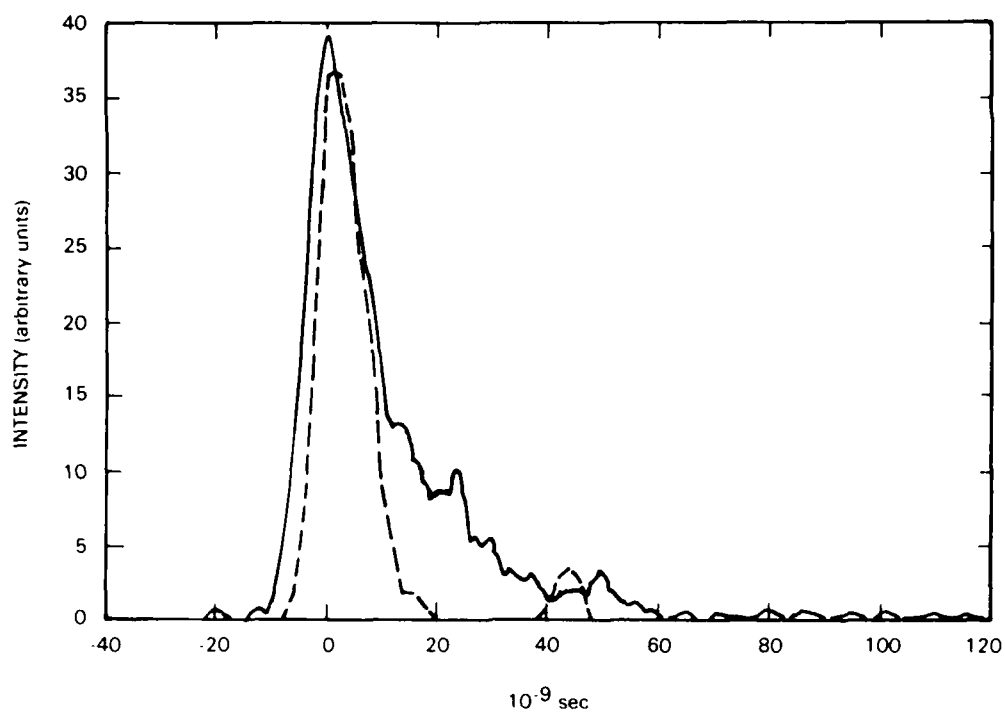


Figure 5. Pulse shapes of the 193-nm pump laser (dashed line) and the output at 455 nm (solid line).

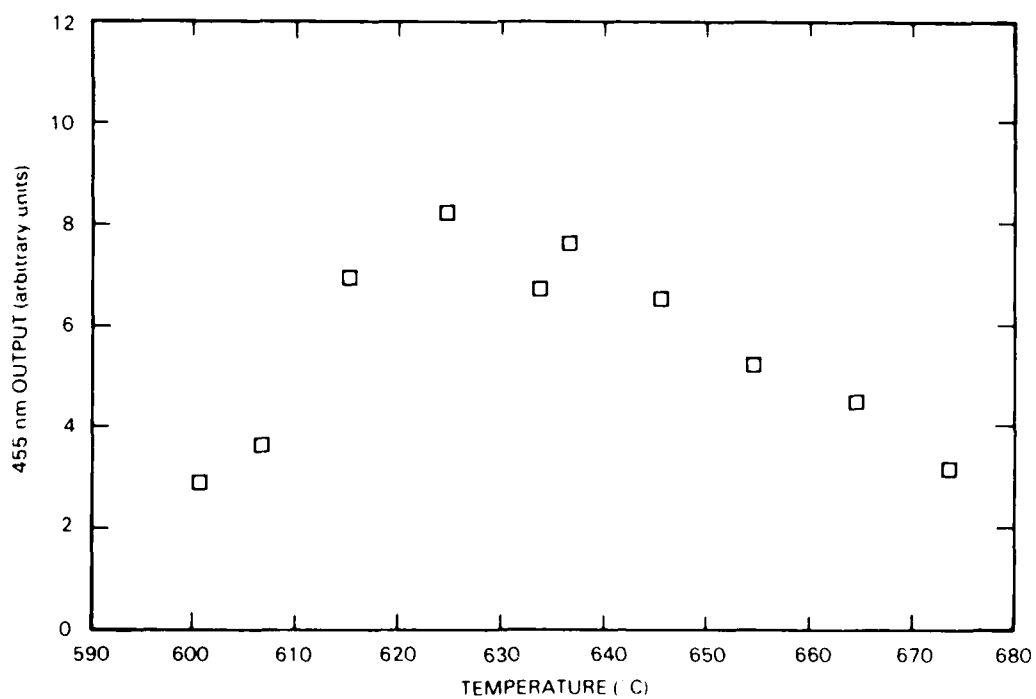


Figure 6. Relative output at 455-nm for constant pump intensity as a function of side arm temperature with the main cell kept at 753 C

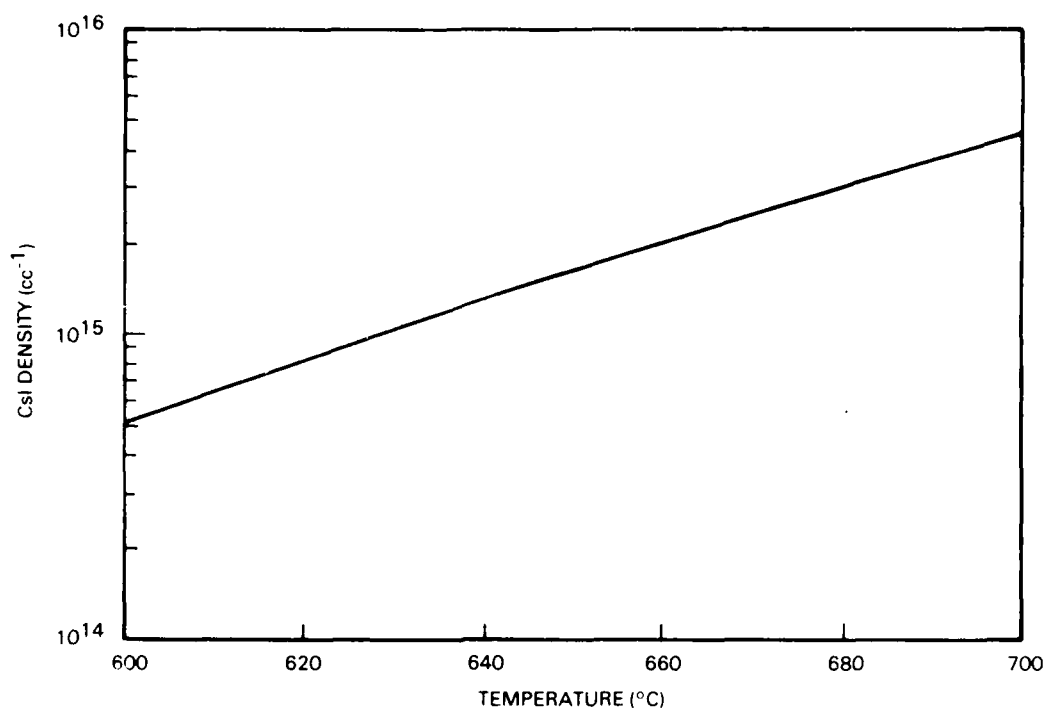


Figure 7. Number density of saturated cesium iodide vapor calculated from data in reference 8.

Energy in/out curves are shown in figure 8 for 917 nm and 876 nm and in figure 9 for 455 nm. The ordinate scales in these two figures should be used more as a relative indication of output rather than an absolute one. The absolute accuracy is probably no better than 1 or 2 decades (see section 2). A slope greater than unity in such a log/log plot is an indication of super-radiance and is evident in the middle energy portion of the 917-nm data and the low energy portion of the 876-nm data. At energies in excess of 1 or 2 mJ, saturation is apparent due to decreased efficiency in absorption of pump light. The output at 455 nm is significantly weaker than that observed for the two measured infrared lines and is seen to vary linearly with pump intensity over 2 orders of magnitude. The addition of a highly reflective mirror at 455 nm (figure 2) made only a marginal improvement.

A study was made of potential nonlinear optical crystals for parametric amplification of the 455-nm output from the CsI cell. Calculations were performed for laser pump wavelengths of 355 nm from frequency tripled Nd:YAG and 308 nm from XeCl. The idler wavelengths required by these two pumps are 1602 nm and 951 nm respectively. The nonlinear crystal must be reasonably transparent over this range and, of course, be able to phase match the sum frequency analog process with appropriate polarizations:

$$\begin{array}{ccccc} n_{455} & + & n_{\text{idler}} & = & n_{\text{pump}} \\ \hline 455 & & \lambda_{\text{idler}} & & \lambda_{\text{pump}} \end{array}$$

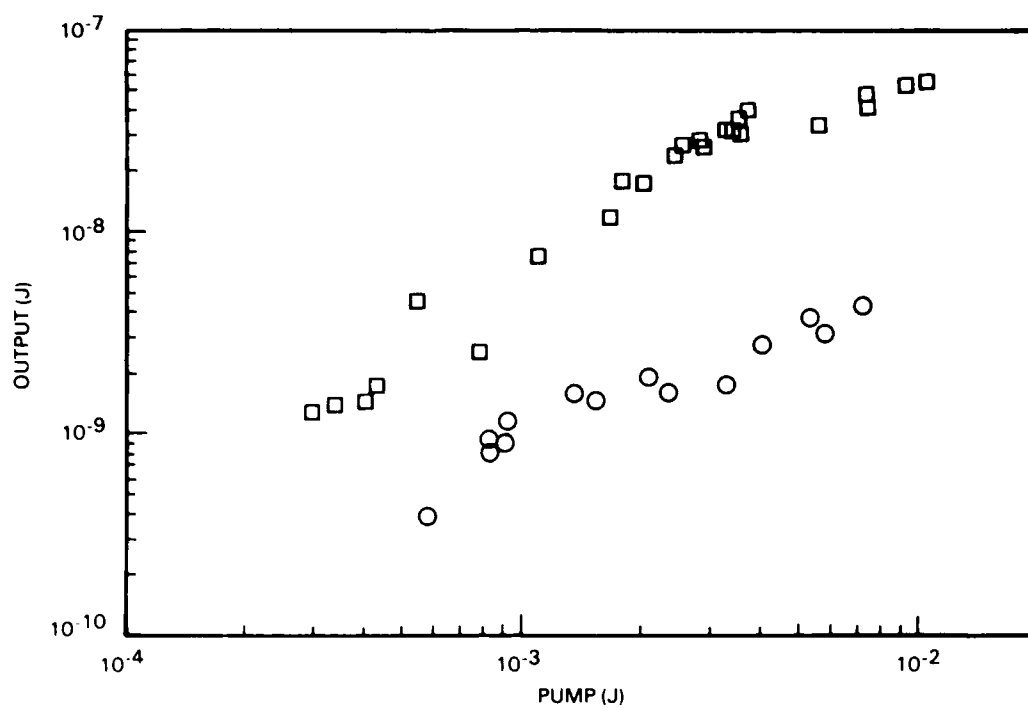


Figure 8. Measured cesium emission at 917 nm and 876 nm as a function of pump energy.

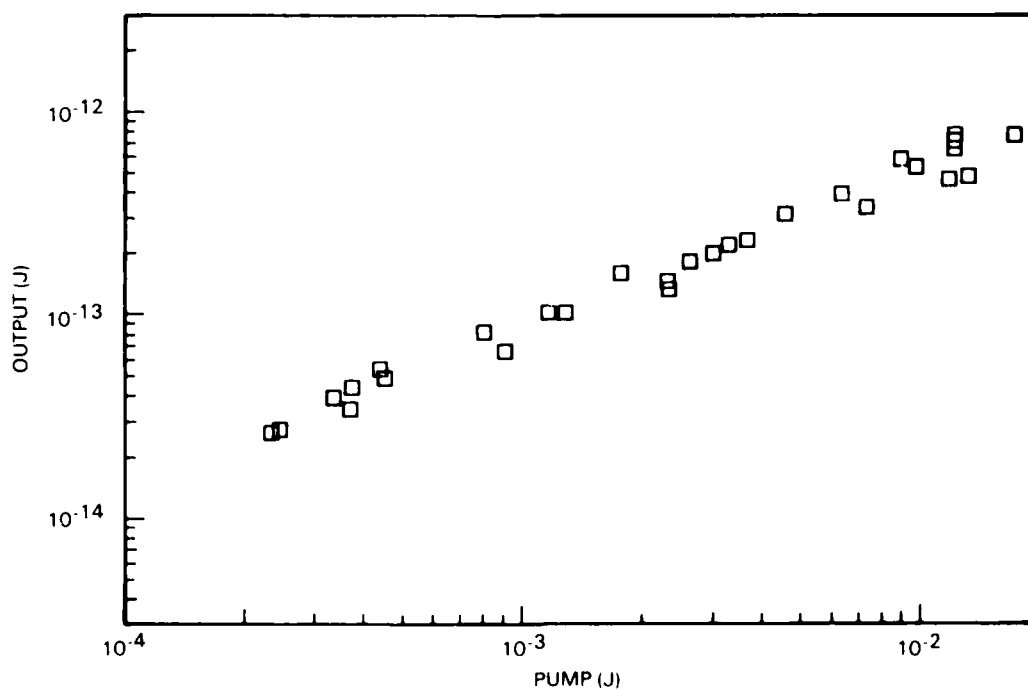


Figure 9. Measured cesium emission at 455 nm as a function of pump energy.

For the ideal case of plane waves, perfect phase matching, no absorption loss, and no pump depletion, the signal at 455 nm will grow according to

$$S_1(x) = S_1(0)\cosh^2(gx/2).$$

where the gain  $g$  is given by

$$g = 1.38 \times 10^{13} \times \left( \frac{2S_3}{n_1 n_2 n_3 \lambda_1 \lambda_2} \right)^{1/2} d_{\text{eff}}$$

in MKS units (reference 10). Here 1, 2, and 3 refer to the signal, idler, and pump respectively.  $S_i$  is optical flux in  $\text{W}/\text{cm}^2$ , and  $d_{\text{eff}}$  is the effective (angle dependent) nonlinear coefficient for this process. The calculated gain for KDP (type I), KD\*P (type II), and LiIO<sub>3</sub> (type I) using 308-nm, 355-nm, and 355-nm pumps respectively is shown in figure 10 as a function of pump intensity. Phase matching angles were calculated using indices of refraction supplied by Cleveland Crystals and the effective nonlinear coefficients were taken from references 10 and 11. Thus, for example, a 5 cm length of KDP pumped with 1  $\text{GW}/\text{cm}^2$  of 308-nm will give an overall parametric amplification of over 3000. It should be noted that with 5 to 10 ns pulses, optical damage is likely to occur at power levels much in excess of 1  $\text{GW}/\text{cm}^2$  for the KDP's (reference 12) and 100  $\text{MW}/\text{cm}^2$  for LiIO<sub>3</sub> (reference 13). Unfortunately, the Csl output at 455-nm was insufficient to attempt this experiment.

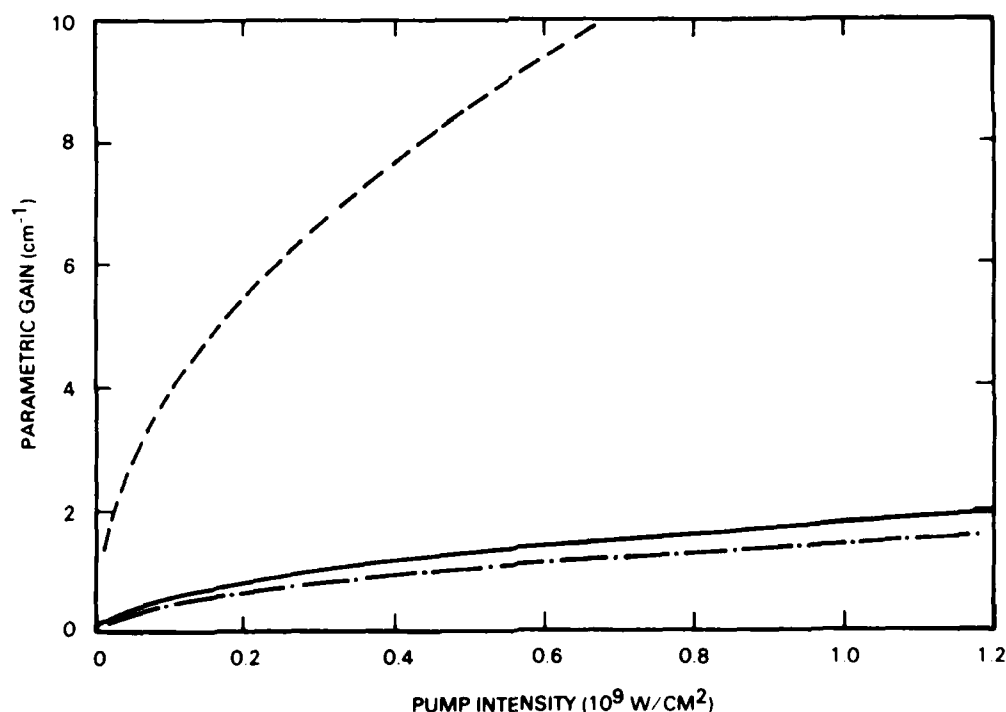


Figure 10. From bottom to top: calculated parametric gain at 455 nm for KD\*P (type II, 355 nm pump), KDP (type I, 308 nm pump), and LiIO<sub>3</sub> (type I, 355 nm pump) as a function of pump power. Phase matching angles are 43, 54, and 41 degrees respectively.

#### 4. CONCLUSION

The results from these experiments were essentially negative. Generating a spectrally narrow and stable laser source by dissociating alkali halides may be potentially useful at certain wavelengths, but is unsuitable in this case. The relatively weak spontaneous fluorescence observed at the second resonance lines 455-nm and 459-nm can be due to three factors: insufficient initial population of the 7P states following dissociation of the parent diatomic Csl, ground state buildup, and/or competition from other lines which deplete the 7P population.

Perhaps some improvement would be possible with a photodissociation source more closely matched to the cesium 7P asymptote near 198-nm, although a large improvement is not likely since the production efficiency is already estimated to be 10-20 percent using the 193-nm pump source (reference 2). The latter two factors are intrinsic problems for operation at this wavelength. It is worth noting that for a potentially useful atomic resonance filter, operation from the ground state is attractive since optical absorption can be made high. However, for the analogous laser system, operation from the ground state is a detrimental factor.

## 5. REFERENCES

1. Marling, J. B., J. Nilsen, L.C. West, and L.L. Wood, "Advanced Blue-Green Electro-Optics: Very Narrow Bandwidth, Wide Field of View Detectors," UCID-17915, 1977
2. Ehrlich, D. J., and R.M. Osgood, "Alkali-Metal Resonance-Line Lasers Based on Photodissociation," Appl. Phys. Lett. 34, 655 (1979)
3. Ehrlich, D. J., and R.M. Osgood, "Metal-Atom Resonance-Line Lasers," IEEE J. Quantum. Electron. QE-16, 257 (1980)
4. Davidovits, P., and D.C. Brodhead, "Ultraviolet Absorption Cross Sections for the Alkali Halide Vapors," J. Chem. Phys. 46, 2968 (1967)
5. Gaydon, A.G., *Dissociation Energies*, Chapman and Hall Ltd., London 1968
6. Moore, C.E., "Atomic Energy Levels Vol. III," National Bureau of Standards Circular 467, 1958
7. Neiger, M., W. Kaesler, and H.-P. Popp, "Inversion of Atomic Resonance Transitions by Electron Collisional Dissociation," Appl. Phys. B 37, 73 (1985)
8. Stull, D.R., "Vapor Pressure of Pure Substances - Inorganic Compounds," Ind. Eng. Chem. 39, 540 (1947)
9. Miller, R.C., and P. Kusch, "Molecular Composition of Alkali Halide Vapors," J. Chem. Phys. 25, 860 (1956)
10. Yariv, A., *Quantum Electronics, 2nd ed.*, John Wiley & Sons, 1975
11. Zernicke, F. and J. E. Midwinter, *Applied Nonlinear Optics*, John Wiley & Sons, 1973
12. "Second Harmonic Generation by Angle Tuning in Tetragonal Phosphates and Arsenates," Data Sheet 706, Quantum Technology, Inc. 1983
13. Nath, G., H. Mehmanesch, and M. Gsanger, "Efficient Conversion of a RubyLaser to 0.347  $\mu$  in Low Loss Lithium Iodate," A. Phys. Lett. 17, 286 (1970)

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